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Geochemistry of Coexisting Biotite and Muscovite of Portuguese Peraluminous Granitic Differentiation Series

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Abstract

In nine Portugese peraluminous Hercynian granitic series of differentiation, Cr, V, Sc and Ba decrease, whereas Nb, Zn, Sn, Li, Rb and Cs increase in the sequence of micas crystallization. Commonly equilibrium was not attained for trace elements between coexisting primary biotite and muscovite. Correlations of Cr, V, Nb, Li, Rb and Cs were found for biotite-muscovite pairs. The same correlation has a different slope in distinct series due to distinct degree of fractional crystallization, but also to solid-liquid reequilibration during late-magmatic evolution, as suggested by regression lines, which do not generally pass through the origin. Most trace elements partition in favour of biotite, while Sn, Sc, Sr and Ba prefer muscovite. These micas probably crystallized simultaneously as suggested by intergrowths. The partition ratio for Cs is one series is similar to that found experimentally.

Key words: biotite, muscovite, equilibrium, direquilibrium, granitic differentiation series

Introduction

Micas are important in the evolution of peraluminous granitic series. Major and trace elements of micas give good information during the evolution of granitic magmas (e.g.,

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Neiva 1973, 1977, 1993, 1998; Albuquerque 1975; Neiva and Gomes 1991; Neiva et al. 1987; Hall et al. 1993; Silva and Neiva 1990, 2000; Tischendorf et al. 2001). The crystal chemistry of coexisting biotite and muscovite from peraluminous granites seems to be a good indicator of chemical events during crystallization, as shown by Brigatti et al. (2000).

Partition coefficients are dependent on composition of minerals, temperature, and the degree of attainment of equilibrium (Joliff et al. 1992). Experimental data of trace element partitioning among biotite, muscovite and peraluminous silicic melt are scarce (Icenhower and London 1995). Calculated distribution coefficients for biotite-muscovite pairs are scarce, because equilibrium must be found (e.g., Albuquerque 1975, Bea et al. 1994). They must be compared with those derived from crystal melt distribution coefficients. Discrepances may be due to changes in the geochemical behaviour of trace elements and to the presence of accessory phases.

Trace element microanalyses of single crystals of micas in thin sections were determined by laser ablation coupled to an ICP-mass spectrometer (Bea et al. 1994) and microprobe techniques (Icenhower and London 1995). These methods are the best to be applied, because inclusions can have an important effect in trace element contents of micas (Bea et al. 1993). However bulk analyses of micas will retain their values for petrologic interpretations, if trace elements concentrated in inclusions identified microscopically are not taken into account (e.g., Neiva 1998; Tischendorf et al. 2001).

The evolution of trace elements in separated Fe^{2+}-biotite, siderophyllite and primary muscovite and equilibrium-disequilibrium for trace elements between coexisting micas of nine peraluminous Hercynian granitic series of differentiation from northern and central Portugal are studied. The preference of trace elements either for biotite or muscovite are discussed and compared with studies of coexisting micas from granitic rocks. A few partition ratios are compared with those determined experimentally between biotite and muscovite from peraluminous melt (Icenhower and London 1995). The importance of micas in the behaviour of trace elements of granitic rocks during diffrentiation is also presented.

Occurrence of micas

The micas occur in peraluminous granitic rocks from eight Portuguese areas (Fig. 1) already studied petrologically and chemically by us. Two-mica granite has similar amounts of both micas, but biotite-muscovite granite contains more biotite than muscovite, while muscovite-biotite granite has more muscovite than biotite. The rocks range: 1a) from biotite-hornblende tonalite to biotite-muscovite granite at Rebordelo and 1b) from muscovite-biotite granite to muscovite granite at Ervedosa (Gomes 1996, Gomes and Neiva 2002) from two-mica granite to muscovite granite at Jales (Neiva and Gomes 1991); 3) from muscovite-biotite granite to muscovite granite at Alijó-Sanfins (Neiva 1973); 4) from two-mica granite to muscovite-biotite granite at Torrão (Neiva 1998); 5) four muscovite-biotite granites at Paredes da Beira-Penedono (Silva and Neiva 1990); 6) from biotite-muscovite granodiorite to muscovite-biotite granite porphyry at Serra da Estrela (Neiva et al. 1987); 7) from biotite granite to biotite-muscovite granite at Carregal do Sal-Nelas-Lagares da Beira (Silva 1995, Silva and Neiva 2000); 8) from biotite-muscovite granite to muscovite-biotite granite at Penamacor-Monsanto (Neiva and Campos 1992). Not all of the granitic rock contain coexisting micas.

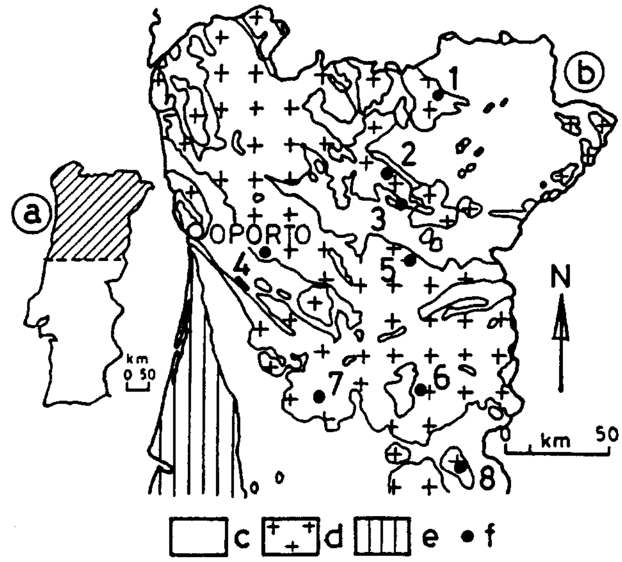

Fig. 1a. Location of Fig. 1b on the map of Portugal. b – Location of the selected areas; c – Pre-ordovician metamorphic complexes, Paleozoic, some igneous and ultrabasic rocks; d – Hercynian granitic rocks; e – Mesozoic and Cenozoic sedimentary rocks;
f – Areas: 1 – Rebordelo-Ervedosa; 2 – Jales; 3 – Alijó-Sanfins; 4 – Torrão; 5 – Paredes da Beira-Penedono; 6 – Serra da Estrela; 7 – Carregal do Sal-Nelas-Lagares da Beira; 8 – Penamacor-Monsanto.

The granitic rocks consist of quartz, K-feldspar, plagioclase, biotite, rare chlorite, muscovite, apatite, monazite, zircon, ilmenite and rutile. Tourmaline occurs in granitic rocks from Rebordelo, Ervedosa, Jales, Alijó-Sanfins, Serra da Estrela and Carregal do Sal-Nelas-Lagares da Beira. Andalusite and sillimanite were found in granites from Ervedosa, Jales, Torrão and Penamacor-Monsanto and sillimanite also occurs in granites and the granite porphyry from Serra da Estrela. Sphene was only found at Rebordelo, while garnet occurs only in the two-mica granite from Jales and cordierite appears only in the biotite granite from Carregal do Sal.

Average chemical compositions and trace elements of these granitic rocks are given in Tables 1, 2 and 3. The Al saturation index given by the molecular ratio Al_2O_3/(CaO+Na_2O+K_2O) ranges between 1.02 and 1.39.

Analytical methods

Biotite and muscovite were separated by magnetic separator and heavy liquids. A purity of about 99.8% was estimated by optical examination. The main contaminants are zircon, monazite and apatite. Several samples were selected from each granite. The trace elements were measured in the separated biotite and muscovite from each sample.

The major and trace elements of granitic rocks and trace elements of micas were mainly determined by X-ray fluorescence at Manchester University, U.K., using the method of Brown et al. (1973). Precisions for major elements and Rb were better than 1% and for trace elements about ±4%. The major elements of micas were determined using the modified Cambridge Geoscan elec-

Table 1. Average chemical analyses in wt.% and trace elements in ppm of peraluminous granitic rocks from northern Portugal.

Area	Rebordelo		Ervedosa		Jales			Alijó-Sanfins			
	1 a–c	1 a–d	1 b–c	1 b–d	2-c	2-d	2-e	3-c	3-d	3-e	3-f
SiO_2	67.89	69.82	72.40	73.88	72.58	73.89	74.96	71.88	71.98	72.25	73.28
TiO_2	0.66	0.48	0.25	0.16	0.24	0.14	0.04	0.26	0.22	0.11	0.07
Al_2O_3	15.23	15.34	14.86	14.71	15.37	14.64	14.80	15.33	15.58	15.61	15.27
Fe_2O_3	0.30	0.15	0.27	0.34	0.33	0.33	0.03	0.23	0.36	0.42	0.21
FeO	3.44	2.38	1.19	0.63	1.11	0.77	0.52	1.19	0.99	0.68	0.58
MnO	0.20	0.11	0.10	0.10	0.02	0.04	0.02	0.02	0.02	0.02	0.03
MgO	1.42	0.96	0.49	0.26	0.54	0.33	0.20	0.13	0.14	0.05	0.04
CaO	2.29	1.57	0.71	0.49	0.70	0.50	0.43	0.84	0.72	0.66	0.56
Na_2O	2.92	3.38	3.24	3.48	3.11	3.28	3.59	3.84	3.89	3.64	3.84
K_2O	4.52	4.85	5.46	5.15	4.90	4.87	4.33	5.13	4.62	4.84	4.61
P_2O_5	0.25	0.27	0.30	0.22	0.28	0.35	0.27	0.29	0.28	0.32	0.30
H_2O+	0.47	0.50	0.64	0.58	0.73	0.83	0.51	0.82	0.98	0.99	1.04
H_2O-	0.26	0.10	0.17	0.04	0.14	0.12	0.18	0.21	0.14	0.52	0.34
Total	99.85	99.91	100.08	100.04	100.05	100.09	99.88	100.17	99.92	100.11	100.17
Cr	52	35	25	12	8	4	*	10	7	6	4
V	68	38	12	4	n.d.	n.d.	n.d.	17	14	9	6
Nb	19	19	19	13	14	16	12	n.d.	n.d.	n.d.	n.d.
Zn	69	58	54	45	58	47	32	n.d.	n.d.	n.d.	n.d.
Sn	31	34	31	30	5	13	7	20	22	36	42
Li	152	154	107	77	160	211	143	355	408	418	445
Ni	13	7	9	*	11	11	10	9	7	3	2
Sc	11	4	8	*	*	*	*	*	*	*	*
Y	59	53	46	43	16	21	8	9	13	9	17
Sr	194	144	78	43	86	61	37	77	69	48	32
Ba	630	460	286	120	245	153	103	420	306	170	147
Rb	271	288	291	346	373	416	396	400	447	563	627
Cs	20	12	17	18	*	15	*	25	29	73	110
A/CNK	1.10	1.12	1.18	1.21	1.31	1.27	1.30	1.14	1.23	1.26	1.24

1 a–c – fine- to medium-grained porphyritic biotite granodiorite; 1 a–d – medium-grained porphyritic biotite-muscovite granite; 1 b–c – medium-grained slightly porphyritic muscovite-biotite granite; 1 b–d – medium- to fine-grained muscovite-biotite granite;

2-c – medium- to coarse-grained porphyritic seriate two-mica granite; 2-d – coarse-grained slightly porphyritic muscovite-biotite granite; 2-e – fine-grained muscovite granite;

3-c – coarse-grained gneissose porphyritic muscovite-biotite granite; 3-d – fine-grained gneissose porphyritic muscovite-biotite granites; 3-e – medium-grained muscovite-biotite granite; 3-f – fine-grained feebly porphyritic muscovite-biotite granite.

* Below de limit of sensitivity; n.d. – not determiend; A/CNK – molecular $Al_2O_3/(CaO+Na_2O+K_2O)$.

tron microprobe equipped with a Link Analytical energy-dispersive X-ray analyser at Manchester University.

FeO of rocks and micas were determined by titration with a standardised potassium permanganate solution, and H_2O+ was determined using a Penfield tube; both with the precision of ±1%. Total Fe_2O_3 of biotites and Li of rocks and micas were determined by atomic absorption, while C1 and F of micas were measured with selective ion electrodes, but all with the precision of ±2%.

However the major elements of granites and their micas from Alijó-Sanfins were determined by XRF, using the method of Norrish and Hutton (1969). The trace elements Cr, V, Sn, Li, Ni, Sr, Ba, Rb and Cs of rocks and micas and Nb, Zn, Sc and Y of rocks were measured by emission spec-

Table 2. Average chemical analyses in wt.% and trace elements in ppm of granites from northern Portugal.

Area	Torrão		Paredes da Beira-Penedono			
	4-c	4-d	5-a	5-b	5-c	5-d
SiO_2	71.51	72.18	72.34	72.82	73.56	73.90
TiO_2	0.36	0.31	0.21	0.19	0.21	0.13
Al_2O_3	15.09	14.81	15.33	15.25	14.66	15.11
Fe_2O_3	0.12	0.12	0.48	0.39	0.45	0.27
FeO	1.64	1.47	0.79	0.91	0.94	0.91
MnO	0.10	0.10	0.05	0.05	0.05	0.05
MgO	0.70	0.57	0.40	0.38	0.37	0.29
CaO	0.98	0.81	0.46	0.43	0.53	0.35
Na_2O	3.65	3.68	2.65	3.03	3.16	3.00
K_2O	4.81	5.11	5.42	5.08	5.05	4.96
P_2O_5	0.25	0.32	0.34	0.32	0.40	0.26
H_2O+	0.70	0.47	1.19	0.89	0.52	0.40
H_2O–	0.07	0.04	0.26	0.26	0.09	0.28
Total	99.98	99.99	99.92	100.00	99.99	99.91
Cr	17	11	21	23	23	21
V	24	17	8	11	13	8
Nb	16	25	20	24	20	22
Zn	78	67	58	71	91	64
Sn	*	7	*	5	*	*
Li	67	82	108	174	234	201
Ni	*	*	3	4	4	4
Sc	*	*	6	5	*	5
Y	40	44	19	21	20	20
Sr	215	123	84	62	56	46
Ba	599	300	229	143	124	83
Rb	260	374	305	383	484	484
Cs	n.d.	n.d.	7	18	31	11
A/CNK	1.16	1.14	1.39	1.35	1.26	1.38

4-c – medium- to fine-grained two-mica granite; 4-d – medium-grained porphyritic muscovite-biotite granite;
5-a – fine-grained gneissose porphyritic muscovite-biotite granite; 5-b – medium-grained gneissose seriate muscovite-biotite granite; 5-c – medium-grained porphyritic muscovite-biotite granite; 5-d – medium-grained porphyritic muscovite-biotite granite.
*Below the limit of sensitivity; n.d. – not determined.

troscopy with a precision of ±20–25% at the University of Cambridge, U.K. (Neiva 1973). Nb, Zn, Sc and Y of micas from Alijó-Sanfins were determined with an Associated Electrical Industries Limited MS7 spark source mass spectrograph with a precision of ±7% and the limit of detection of 0.01 ppm at the University of Manchester (Neiva 1973).

Table 3. Average chemical analyses in wt.% and trace elements in ppm of granitic rocks from central Portugal.

Area	Serra da Estrela			Carregal do Sal – Nelas – Lagares da Beira		Penamacor – Monsanto	
	6-c	6-d	6-e	7-c	7-d	8-c	8-d
SiO_2	74.32	73.91	75.97	71.21	73.35	73.52	75.04
TiO_2	0.29	0.12	0.08	0.48	0.29	0.36	0.22
Al_2O_3	13.55	14.58	13.28	14.07	14.09	12.50	12.41
Fe_2O_3	0.22	0.49	0.52	0.27	0.10	0.36	0.36
FeO	1.55	0.61	0.50	2.36	1.64	1.75	1.03
MnO	0.06	0.06	0.06	0.03	0.04	0.03	0.02
MgO	0.44	0.24	0.21	0.76	0.53	1.34	1.17
CaO	0.71	0.47	0.30	1.20	0.97	0.74	0.55
Na_2O	3.24	3.19	3.20	2.89	3.01	3.70	3.85
K_2O	4.70	4.98	4.57	5.12	5.06	4.49	4.41
P_2O_5	0.28	0.24	0.22	0.27	0.20	0.56	0.41
H_2O+	0.59	0.99	1.01	0.94	0.58	0.65	0.50
$H_2O–$	0.05	0.17	0.14	0.05	0.04	0.08	0.02
Total	100.00	100.05	100.06	99.65	99.90	100.08	99.99
Cr	24	22	21	9	7	33	27
V	n.d.	n.d.	n.d.	31	17	7	3
Nb	10	6	5	25	31	9	12
Zn	80	54	66	66	64	111	122
Sn	*	*	*	22	36	*	11
Li	118	64	67	140	124	134	162
Ni	7	10	9	17	18	7	6
Sc	6	5	3	5	*	4	4
Y	45	45	36	43	31	26	27
Sr	76	74	19	87	94	55	21
Ba	202	146	5	302	368	214	75
Rb	344	336	332	351	355	271	364
Cs	5	5	*	20	18	n.d.	n.d.
A/CNK	1.16	1.27	1.24	1.13	1.16	1.02	1.03

6-c – coarse-grained porphyritic biotite-muscovite granite; 6-d – fine- to medium-grained slightly porphyritic muscovite-biotite granite; 6-e – granite porphyry;
7-c – coarse- to very coarse-grained porphyritic biotite granite; 7-d – medium- to coarse-grained porphyritic biotite-muscovite granite;
8-c – coarse- to medium-grained porphyritic biotite-muscovite granite; 8-d – coarse-grained porphyritic muscovite-biotite granite.
n.d. – not determined; * below the limit of sensitivity.

Chemical composition of micas

Major and trace element analyses of coexisting biotite and muscovite from Portuguese peraluminous granitic differentiation series are given in Tables 4, 5, 6, 7, 8 and 9. The biotites are mainly Fe^{2+}-biotite (Figs. 2a, b), but a few from Paredes da Beira-Penedono and Serra da Estrela have a composition ranging from Fe^{2+}-biotite to siderophyllite (Fig. 2b) according to the nomenclature of Foster (1960). The muscovites correspond to compositions of primary muscovite (Miller et al. 1981; Monier et al. 1984) as shown in Fig. 3 and contain a significant celadonitic component.

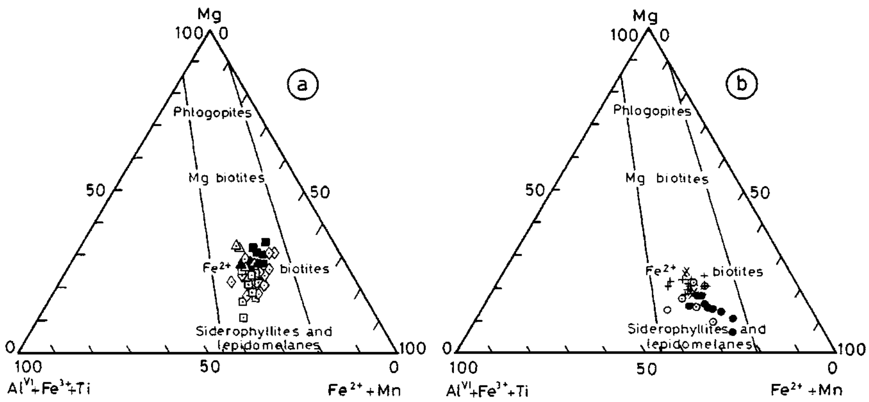

Fig. 2. Compositions of biotite from Portuguese granitic differentiation series.
a. ■ Rebordelo, ◇ Ervedosa, ⊡ Jales, ▲ Alijó-Sanfins, △ Torrão;
b. ⊙ Paredes da Beira-Penedono, ● Serra da Estrela, + Carregal do Sal-Nelas-Lagares da Beira, × Penamacor-Monsanto.

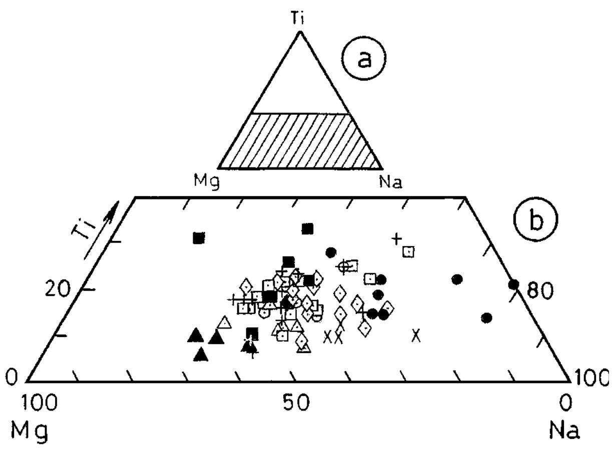

Fig. 3. Compositions of muscovite from Portuguese granitic differentiation series. Symbols as in Fig. 2.

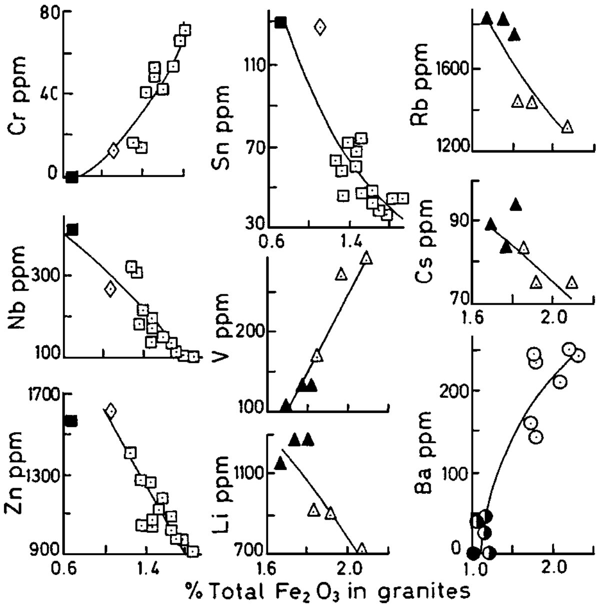

Fig. 4. Variation diagrams of trace elements of biotite from Portuguese granitic series. Symbols: ⊡ two-mica granite, ◇ muscovite-biotite granite, ■ muscovite granite from Jales; △ two-mica granite, ▲ muscovite-biotite granite from Torrão; ⊙ biotite-muscovite granite, ◑ muscovite-biotite granite, ● granite porphyry from Serra da Estrela.

Data of bulk trace elements were determined on mineral separates. However contents of Zr, U, Th and REE (mainly La, Ce, Nd) are constituents of inclusions and therefore are not considered. The degree of fractionation of granitic melt controls the chemical composition of micas studied, particularly the behaviour of trace elements. Generally Cr, V, Sc, Ba contents decrease and Nb, Zn, Sn, Li, Rb, Cs contents increase in both biotite and muscovite, but Ni content also decreases and Y content increases in biotite with decreasing total Fe_2O_3 in the granitic rocks within each Portuguese series studied. The behaviours of some trace elements are shown, e.g., Cr, Nb, Zn and Sn in biotite from the Jales series; V, Li, Rb and Cs in biotite from the Torrão series and Ba in biotite from the Serra da Estrela series (Fig. 4); Cr, Nb, Zn, Sn and Ba in muscovite from Jales series; V, Li, Rb and Cs in muscovite from Torrão series (Fig. 5).

Equilibrium-disequilibrium between coexisting micas

The correlations between coexisting biotite and muscovite concentrations were verified through the Pearson's coefficients. They are significant with a probability of >0.95. Correlations between coexisting micas were found for ten trace elements in a variable number of differentiation series: Cr in series 1a, 2, 4 and 6; V in series 1a and 4; Nb in

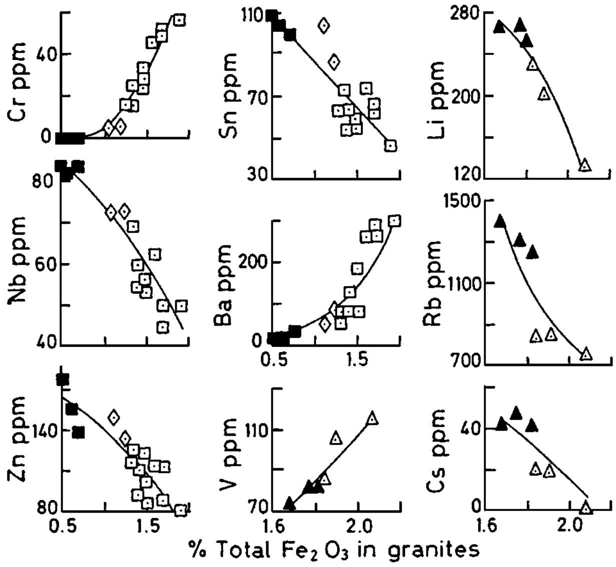

Fig. 5. Variation diagrams of trace elements of muscovite from Portuguese granitic series. Symbols as in Fig. 4.

series 1a and 2; Cs in series 2, 3 and 6; Li in series 1a, 1b, 2, 4 and 7; Rb in series 1a, 1b, 2, 4 and 7 (Fig. 6); Zn in series 2; Sn in series 6; Y in series 4; Ba in series 4 (Fig. 7). So the series 2 (Jales) and series 4 (Torrão) have the highest number of correlations for trace elements between coexisting micas (6 for each series) followed by the series 1a (Rebordelo) with five trace elements.

Only the distribution of Cr in the biotite/muscovite pairs of series 2 and 4 and of Cs in these pairs for series 6 pass through the origin (Fig. 6) and represent equilibrium between coexisting micas for these trace elements. Biotite and muscovite occur in independent flakes, but are also intergrown in other flakes, as observed microscopically in all samples suggesting that they grew simultaneously.

In most of the series, the regression lines do not pass through the origin (Figs. 6 and 7), which can be related to solid-liquid reequilibration during the late-magmatic evolution. Furthermore, the variations in composition and temperature are not enough great within each series, except in series 1a, to explain that most of the correlations do not pass through the origin (Icenhower and London 1995). Disequilibrium predominates over equilibrium on the distribution of trace elements between coexisting micas. Trace elements were determined on mica separates, but this technique may not be responsible for the general disequilibrium found, because a similar procedure was applied in Viseu area, central Portugal and equilibrium was found between coexisting micas from two granitic suites (Neves 1997).

Generally the same correlation between coexisting micas shows distinct slopes for different differentiation series (Fig. 6), which would be attributed to different degrees of fractional crystallization if the regression lines were passing through the origin. Therefore they are also probably due to solid-liquid reequilibration during late-magmatic evolution.

Distribution coefficients

Distribution coefficients can only be calculated if a linear correlation was found for a trace element between two coexisting minerals. If the regression line does not pass through the origin, the distribution coefficient is expressed as a linear funktion. If the regression lines pass through the origin, partition ratios are calculated dividing the concentration of any trace element in biotite by its content in the coexisting muscovite (Beattie et al. 1993). $D(M)^{Bt/Ms}$ where D, Bt, Ms and M represent the partition ratio, biotite, muscovite and the trace element of interest respectively. Calculated distribution coefficients for trace elements analyzed on separates of coexisting biotite and muscovite from different Portuguese granitic series are given in Table 10. Partition ratios could only be calculated for Cr of series 2 (Jales) and series 4 (Torrão) and for Cs of series 6 (Serra da Estrela), because only for them regression lines pass through the origin.

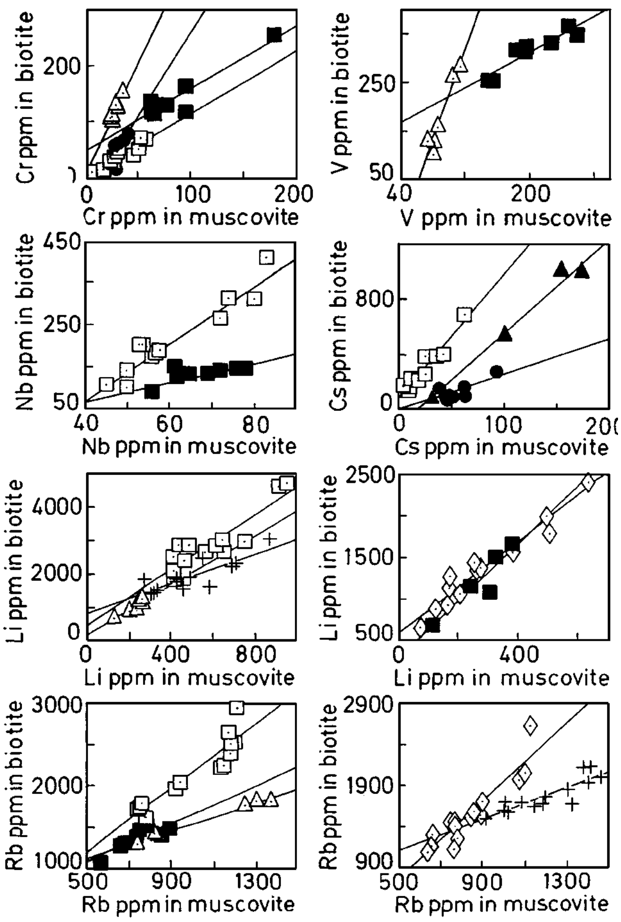

Fig. 6. Correlations of Cr, V, Nb, Cs, Li and Rb between coexisting biotite and muscovite from some Portuguese granitic differentiation series.
Symbols: ■ Rebordelo – 1a, ◇ Ervedosa – 1b, ⊡ Jales – 2, ▲ Alijó-Sanfins – 3, △ Torrão – 4, ● Serra da Estrela – 6, + Carregal do Sal-Nelas-Lagares da Beira – 7. Numbers of the series.

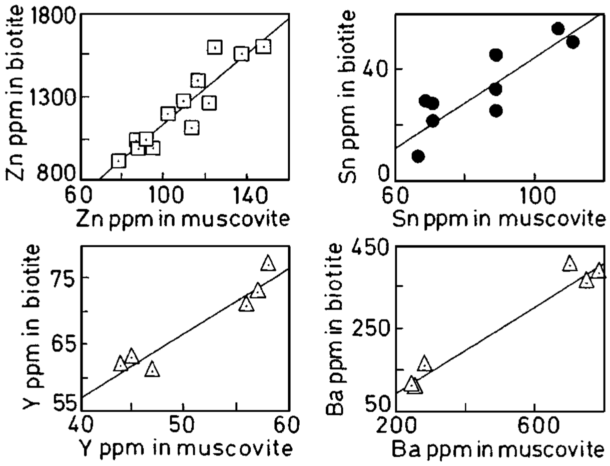

Fig. 7. Correlations of Zn, Sn, Y and Ba between coexisting micas from Portuguese granitic differentiation series.
Symbols: ⊡ Jales – 2, ● Serra da Estrela – 6, △ Torrão – 4. Numbers of the series.

Table 4. Average chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of peraluminous granitic rocks from Rebordelo and Ervedosa, northern Portugal.

	Rebordelo				Ervedosa			
	Biotite 1 a-c	Muscovite 1 a-c	Biotite 1 a-d	Muscovite 1 a-d	Biotite 1b-c	Muscovite 1 b-c	Biotite 1 b-d	Muscovite 1 b-d
SiO_2	36.00	46.31	35.42	45.46	35.40	45.87	36.13	46.59
TiO_2	2.94	1.23	2.55	1.10	2.69	0.63	2.18	1.01
Al_2O_3	18.05	33.46	18.82	34.75	19.05	35.32	20.48	34.49
Fe_2O_3	1.71	0.47	2.06	0.43	2.36	0.50	3.33	0.54
FeO	20.25	0.98	20.56	0.88	21.09	0.84	20.22	0.95
MnO	0.27	0.01	0.23	0.03	0.19	0.02	0.23	–
MgO	7.24	1.24	7.30	0.80	6.38	0.71	4.44	0.85
CaO	0.09	0.01	0.02	0.01	0.03	0.03	0.03	–
Na_2O	0.20	0.43	0.39	0.62	0.45	0.63	0.46	0.76
K_2O	9.63	10.50	9.37	10.28	9.32	10.34	9.11	10.25
Cl	0.06	0.03	0.05	0.03	0.05	0.03	0.04	0.03
F	0.35	0.13	0.37	0.17	0.33	0.14	0.45	0.25
	96.79	94.80	97.14	94.56	97.34	95.06	97.10	95.72
O ≡ Cl	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
O ≡ F	0.15	0.05	0.16	0.07	0.14	0.06	0.19	0.11
Total	96.63	94.74	96.97	94.48	97.19	94.99	96.90	95.60
Cr	188	137	134	71	101	31	59	19
V	360	255	301	185	175	67	104	51
Nb	107	59	852	70	207	76	311	103
Zn	421	51	576	90	851	73	1170	117
Sn	130	159	145	165	137	113	258	132
Li	1100	305	1266	273	1378	266	2313	400
Ni	92	10	77	14	67	11	76	16
Sc	37	58	27	55	10	17	3	13
Y	71	50	72	45	85	48	90	56
Sr	23	35	21	30	20	24	21	21
Ba	418	598	81	381	*	181	*	56
Rb	1227	654	1406	773	1379	799	2296	1104
Cs	70	12	103	16	127	16	393	15

Column headings as in Table 1. – not detected; * below the limit of sensitivity.

Table 5. Average chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of peraluminous granitic rocks from Jales, northern Portugal.

	Biotite 2-c	Muscovite 2-c	Biotite 2-d	Muscovite 2-d	Biotite 2-e	Muscovite 2-e
SiO_2	35.93	46.65	36.35	46.07	35.48	46.41
TiO_2	2.58	0.74	2.35	0.51	1.55	0.27
Al_2O_3	19.88	35.15	21.41	34.95	22.37	35.34
Fe_2O_3	1.64	0.27	1.49	0.40	2.83	0.30
FeO	20.41	1.30	20.85	1.60	19.99	1.93
MnO	0.46	0.02	0.53	0.04	1.33	0.11
MgO	5.52	0.77	3.67	0.87	2.37	0.68
CaO	0.01	0.01	0.01	–	–	–
Na_2O	0.09	0.55	0.07	0.56	0.05	0.40
K_2O	9.27	10.12	9.09	10.28	8.93	10.21
Cl	0.03	–	0.02	–	0.03	–
F	0.71	0.36	0.79	0.52	0.54	0.30
	96.53	95.94	96.63	95.80	95.47	95.95
O ≡ Cl	0.01	–	–	–	0.01	–
O ≡ F	0.30	0.15	0.33	0.22	0.23	0.13
Total	96.22	95.79	96.30	95.58	95.23	95.82
Cr	43	34	13	5	*	*
V	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Nb	191	58	270	72	410	83
Zn	1167	103	1595	141	1560	157
Sn	54	60	129	96	130	103
Li	2614	569	4617	910	4731	941
Ni	57	17	25	10	9	19
Sc	8	24	*	16	*	9
Y	64	38	104	9	58	29
Sr	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Ba	159	158	57	65	*	15
Rb	2085	978	2520	1178	2933	1281
Cs	253	16	696	59	770	38

Column headings as in Table 1. – not detected; * below the limit of sensitivity; n.d. - not determined.

Most trace elements have a higher concentration in biotite than in coexisting muscovite (Tables 4, 5, 6, 7, 8 and 9), but Sn, Sc and Ba show preference for muscovite (e.g., Albuquerque 1975), which cannot be the result of late muscovite crystallization from an enriched residual melt, because this melt would have been impoverished in Sc and Ba as shown by the decrease in Ba content in biotite from Serra da Estrela series (Fig. 4) and muscovite from Jales series (Fig. 5).

Cr, V, Nb, Zn, Sn, Li, Ni and Sc seem likely to substitute in octahedral sites in micas. Cr^{3+} (6.3 nm) and V^{3+} (7.4 nm) will substitute for Fe^{3+} (6.4 nm); Li^{+} (6.8 nm) will replace Mg^{2+} (6.6 nm), but requires a coupled substitution (Foster 1960); Ni^{2+} (6.9 nm) will replace Mg^{2+}; and Zn^{2+} (7.4 nm) and Sc^{2+} (8.1 nm) will substitute for Fe^{2+} (7.4 nm). These trace elements replace major elements which are more abundant in biotite than in muscovite (Tables 4 to 9). Consequently these elements are generally more concentrated in biotite than in coexisting muscovite. Partition ratio for Cr in series 4 is higher by two orders of magnitude than in series 2 (Table 10), but similar to that found by Neves (1997). In biotite/muscovite pairs commonly Cr and V prefer biotite (Albuquerque 1975; Neiva 1977; Neves 1997).

Rare data were found in the literature on Nb and Zn of coexisting biotite and muscovite from granitic rocks, but they prefer biotite rather than coexisting muscovite (Jolliff et al. 1992; Neves 1977) as in this study.

Table 6. Average chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of peraluminous granitic rocks from Alijó-Sanfins, northern Portugal.

	Biotite 3-c	Muscovite 3-c	Biotite 3-d	Muscovite 3-d	Biotite 3-e	Muscovite 3-e	Biotite 3-f	Muscovite 3-f
SiO_2	34.63	45.33	34.09	45.60	35.45	44.56	35.17	45.72
TiO_2	2.41	0.62	2.45	0.42	2.08	0.40	1.70	0.26
Al_2O_3	19.69	34.00	20.62	34.25	20.34	33.26	21.49	33.73
Fe_2O_3	1.52	–	2.29	0.76	2.69	0.48	2.54	–
FeO	19.81	1.75	19.37	1.62	20.83	2.41	19.58	3.45
MnO	0.46	0.04	0.48	0.04	0.49	0.05	0.95	0.10
MgO	6.57	1.97	6.65	1.38	5.01	2.16	4.95	1.49
CaO	1.61	0.63	0.51	–	–	–	–	–
Na_2O	0.22	0.61	0.12	0.71	0.22	0.75	0.19	0.54
K_2O	8.73	10.32	9.04	10.58	9.10	10.08	8.97	10.27
Cl	–	0.06	0.06	0.08	0.09	0.10	0.11	0.11
F	–	0.02	0.14	0.09	0.37	0.31	0.60	0.27
	95.65	95.35	95.82	95.53	96.67	94.56	96.25	95.94
O ≡ Cl	–	0.01	0.01	0.02	0.02	0.02	0.02	0.02
O ≡ F	–	0.01	0.06	0.04	0.16	0.13	0.25	0.11
Total	95.65	95.33	95.75	95.47	96.49	94.41	95.98	95.81
Cr	37	13	48	6	23	5	17	5
V	48	*	48	*	*	*	*	*
Nb	71	29	91	32	133	36	362	39
Zn	242	58	250	186	359	277	543	380
Sn	100	100	100	100	140	207	140	205
Li	3850	500	3900	800	4000	1717	6000	2100
Ni	43	2	37	2	13	2	8	2
Sc	7	15	6	8	2	7	1	2
Y	0.50	0.45	0.60	0.07	0.70	0.05	0.80	0.64
Sr	*	*	*	*	*	*	*	*
Ba	55	100	40	100	40	72	55	60
Rb	2200	900	1360	960	2600	1113	1800	1300
Cs	1000	32	550	100	1000	153	1000	173

Column headings as in Table 1. – not detected; * below the limit of sensitivity.

Li is more concentrated in biotite than in coexisting muscovite (Tables 4 to 9) as found by Carron and Lagache (1972), Albuquerque (1975), Kretz et al. (1989), Jolliff et al. (1992), Bea et al. (1994) and Neves (1997).

Sn in biotite shows a strong resemblance to the behaviour of Fe^{2+}, Fe^{3+} and Ti and is positively correlated with Li (e.g., Hesp 1971; Neiva 1976). Sn in muscovite is negatively correlated with Ti, Mg and positively with F, Li and Rb (Neiva 1977). In general, muscovite has a higher Sn content than coexisting biotite (Neiva 1984), as in the Pedrobernado pluton, central Spain (Bea et al. 1994) and in the granites of Viseu region, central Portugal (Neves 1997). However in the tin-bearing granites from Ervedosa, Sn is preferentially concentrated in biotite compared with coexisting muscovite (Table 4).

The data available on Sr for coexisting biotite and muscovite is only for five series (Tables 4, 7 and 8) and muscovite has a higher Sr content than coexisting biotite. The experimental data on element partitioning among biotite, muscovite and melt (Icenhower and London 1995) and the data on coexisting micas from granitic rocks (Jolliff et al., 1992; Neves 1997) support this finding. Sr has a radius (11.2 nm) closer to that of Ca^{2+} (9.9 nm) than that of K^+ (13.3 nm), but Ca was rarely detected in both micas. However, Y (9.2 nm) probably replaces Ca^{2+}, but it is partitioned preferentially into biotite compared with coexisting muscovite (Tables 4–9).

Table 7. Chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of granites from Torrão and Paredes da Beira – Penedono, northern Portugal.

	Torrão				Paredes da Beira – Penedono							
	Biotite 4-c	Muscovite 4-c	Biotite 4-d	Muscovite 4-d	Biotite 5-a	Muscovite 5-a	Biotite 5-b	Muscovite 5-b	Biotite 5-c	Muscovite 5-c	Biotite 5-d	Muscovite 5-d
SiO_2	35.09	46.07	34.99	45.77	35.39	46.68	35.89	46.21	37.41	46.03	35.90	46.59
TiO_2	2.71	0.69	2.53	0.60	2.53	0.75	2.28	0.50	2.42	0.53	2.44	0.48
Al_2O_3	19.25	35.41	19.57	35.40	19.12	34.53	19.68	33.89	19.61	33.43	19.80	33.21
Fe_2O_3	3.43	0.13	3.47	0.20	3.08	0.68	1.93	0.84	0.27	0.53	0.49	0.71
FeO	17.28	1.14	18.93	1.33	20.73	0.97	21.68	1.76	22.90	2.77	24.88	2.20
MnO	0.22	0.02	0.25	0.06	0.47	–	0.52	–	0.45	–	0.43	–
MgO	7.79	1.35	6.15	1.41	5.25	0.45	4.15	0.59	3.15	0.77	2.32	0.67
CaO	0.16	0.13	0.17	0.15	–	–	–	–	–	0.05	–	–
Na_2O	0.67	0.72	0.57	0.99	0.25	0.54	0.34	0.46	0.28	0.54	0.38	0.47
K_2O	9.52	10.38	9.40	10.12	8.95	10.44	9.15	10.48	9.55	10.39	9.29	10.58
Cl	0.07	–	0.01	–	0.10	–	0.14	–	0.08	–	0.03	–
F	0.54	0.17	0.30	0.26	0.38	0.19	0.38	0.33	0.82	1.11	1.58	0.51
	96.73	96.21	96.34	96.29	96.25	95.23	96.14	95.06	96.94	96.15	97.54	95.42
O ≡ Cl	0.02	–	–	–	0.02	–	0.03	–	0.02	–	0.01	–
O ≡ F	0.23	0.07	0.13	0.11	0.16	0.08	0.16	0.14	0.34	0.47	0.66	0.21
Total	96.48	96.14	96.21	96.18	96.07	95.15	95.95	94.92	96.58	95.68	96.87	95.21
Cr	135	31	103	24	35	32	32	27	52	33	22	24
V	237	103	122	80	41	32	45	18	89	44	24	21
Nb	153	51	233	96	233	120	331	110	244	95	332	112
Zn	774	58	746	76	1451	228	1709	265	1548	172	1779	296
Sn	13	7	12	13	24	62	24	147	39	125	43	89
Li	839	186	1204	262	1231	399	1383	1008	1421	919	1645	912
Ni	53	26	31	22	37	10	39	12	59	17	35	16
Sc	15	19	6	16	*	8	*	14	*	11	8	25
Y	62	45	74	57	36	19	36	28	26	21	42	25
Sr	23	35	20	24	19	20	16	18	15	15	18	16
Ba	383	745	126	265	37	127	12	90	31	77	10	55
Rb	1381	805	1806	1307	1579	854	1704	1272	2257	1415	2476	1473
Cs	77	18	88	42	349	24	410	88	748	97	322	49

Column headings as in Table 2. – not detected; * below the limit of sensitivity.

Table 8. Chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of granites from Serra da Estrela and Carregal do Sal – Nelas – Lagares da Beira, central Portugal.

	Serra da Estrela						Carregal do Sal – Nelas – Lagares da Beira			
	Biotite 6-c	Muscovite 6-c	Biotite 6-d	Muscovite 6-d	Biotite 6-e	Muscovite 6-e	Biotite 7-c	Muscovite 7-c	Biotite 7-d	Muscovite 7-d
SiO_2	36.24	47.29	34.82	47.60	34.32	47.09	35.23	47.02	35.12	46.68
TiO_2	2.60	0.58	2.32	0.43	2.65	0.29	2.68	0.91	2.81	0.73
Al_2O_3	19.10	34.19	19.03	34.78	18.93	34.43	19.35	34.86	19.52	34.17
Fe_2O_3	0.91	0.12	1.24	0.26	0.82	1.01	3.39	0.71	2.98	0.46
FeO	24.17	1.64	25.68	1.30	27.52	1.33	19.60	1.08	20.71	1.45
MnO	0.14	–	0.65	–	0.53	–	0.27	0.03	0.42	0.05
MgO	4.11	0.49	2.93	0.31	1.69	–	5.31	0.52	4.78	1.00
CaO	–	–	–	–	–	–	0.02	0.01	0.03	0.03
Na_2O	0.06	0.64	0.02	0.64	0.08	0.43	0.18	0.64	0.23	0.58
K_2O	9.59	10.52	9.22	10.50	9.17	10.82	9.10	10.19	9.19	10.17
Cl	0.05	–	0.04	–	0.14	–	0.05	0.03	0.05	0.02
F	0.55	0.37	0.69	0.36	0.47	0.31	0.60	0.48	0.49	0.40
	97.52	95.84	96.64	96.18	96.32	95.71	95.78	96.48	96.33	95.74
O ≡ Cl	0.01	–	0.01	–	0.03	–	0.01	0.01	0.01	–
O ≡ F	0.23	0.16	0.29	0.15	0.20	0.13	0.25	0.20	0.21	0.17
Total	97.28	95.68	96.34	96.03	96.09	95.58	95.52	96.27	96.11	95.57
Cr	65	34	29	23	20	24	70	22	113	20
V	n.d.	n.d.	n.d.	n.d.	n.d.	*	212	133	172	98
Nb	178	39	205	43	233	38	121	82	289	95
Zn	1540	217	1843	127	2250	250	581	94	875	109
Sn	32	88	28	77	29	69	191	194	177	202
Li	1718	485	1086	340	1864	306	1478	321	1948	507
Ni	30	18	8	16	*	19	31	24	24	23
Sc	16	24	5	22	*	9	51	81	22	43
Y	54	67	77	62	95	61	99	25	138	47
Sr	9	20	16	22	*	*	n.d.	n.d.	n.d.	n.d.
Ba	206	156	36	132	*	5	123	682	87	453
Rb	1658	1104	1600	997	1863	1081	1703	1006	1786	1218
Cs	134	57	142	47	140	39	126	43	124	41

Column headings as in Table 3. – not detected; n.d. – not determined;* below the limit of sensitivity.

Table 9. Chemical analyses in wt.% and trace elements in ppm of coexisting biotite and muscovite of granitic rocks from Penamacor – Monsanto, central Portugal.

	Biotite 8-c	Muscovite 8-c	Biotite 8-d	Muscovite 8-d
SiO_2	35.92	46.13	35.58	45.90
TiO_2	3.21	0.31	2.39	0.33
Al_2O_3	19.91	35.71	20.42	36.07
Fe_2O_3	2.65	0.09	2.87	0.13
FeO	20.65	0.99	21.99	1.27
MnO	–	–	–	–
MgO	5.86	0.52	4.48	0.51
CaO	–	–	0.09	0.07
Na_2O	–	0.53	0.58	0.82
K_2O	9.50	10.16	8.95	10.25
Cl	0.03	–	0.02	–
F	0.36	0.12	0.40	0.24
	98.09	94.56	97.77	95.59
O ≡ Cl	0.01	–	–	–
O ≡ F	0.15	0.05	0.17	0.10
Total	97.93	94.51	97.60	95.49
Cr	154	50	109	29
V	76	37	38	*
Nb	136	22	230	54
Zn	943	102	1839	178
Sn	*	51	67	145
Li	1122	132	1818	689
Ni	62	9	27	19
Sc	35	26	25	19
Y	66	32	76	62
Sr	n.d.	n.d.	n.d.	n.d.
Ba	268	272	23	51
Rb	1583	790	1977	1505
Cs	n.d.	n.d.	n.d.	n.d.

Column headings as in Table 3. – not detected; * below the limit of sensitivity; n.d. – not determined.

Ba^{3+} (13.4 nm), Rb^+ (14.8 nm) and Cs^+ (16.7 nm) mainly replace K^+ (13.3 nm) in both biotite and muscovite. So they will be accommodated in the interlayer site, which will be smaller in muscovite than in biotite (Shearer et al. 1986). Consequently these elements, particular the larger ones, would be expected to favour biotite as found in this study and as has been documented by other studies of coexisting micas from granitic rocks (Albuquerque 1975; Mittlefehlt and Miller 1983; Shearer et al. 1986; Jolliff et al. 1992; Hall et al. 1993; Bea et al. 1994; Neves 1997) as well as by experimental studies (Icenhower and London 1995). The partition ratio for Cs of series 6 (Table 10) is similar to that found experimentally (Icenhower and London 1995), but lower than that found in Viseu area (Neves 1997). However Ba favours muscovite rather than biotite (Tables 4–9) as found by Albuquerque (1975), Lee et al. (1981), Kretz et al. (1989), Jolliff et al. (1992) and Bea et al. (1994), but experimental data shows the preference of Ba for biotite (Icenhower and London 1995).

Relationships among biotites, muscovites and host rocks

In granitic rocks, micas retain most of Cr, V, Zn, Y, Ni and Li contents. Commonly biotite has higher contents of these trace elements than coexisting muscovite (Tables 4 to

Table 10. Calculated average partition coefficient of trace elements between biotite and muscovite from Portuguese granitic series of differentiation

	Rebordelo	Ervedosa	Jales	Alijó-Sanfins	Torrão	Serra da Estrela	Carregal do Sal
	1a	1b	2	3	4	6	7
Cr	$50+1.1/Cr_{mus}$		2.0		4.4	$-37+2.9/Cr_{mus}$	
V	$130+0.92/V_{mus}$				$-251+4.7/V_{mus}$		
Nb	$-30+2.4/Nb_{mus}$		$-204+6.8/Nb_{mus}$				
Zn			$66+11/Zn_{mus}$				
Sn						$-38+0.83/Sn_{mus}$	
Li	$291+3.5/Li_{mus}$	$582+2.8/Li_{mus}$	$467+4.1/Li_{mus}$		$197+3.7/Li_{mus}$		$817+2.2/Li_{mus}$
Y					$17+0.99/Y_{mus}$		
Ba					$-13+0.53/Ba_{mus}$		
Rb	$506+1.2/Rb_{mus}$	$-411+2.4/Rb_{mus}$	$222+1.9/Rb_{mus}$		$708+0.84/Rb_{mus}$		$621+0.96/Rb_{mus}$
Cs			$94+8.9/Cs_{musc}$	$-113+6.8/Cs_{musc}$		2,8	
n	8	16	13	4	6	10	14

9). The amount of biotite decreases, while that of muscovite increases in each differentiation series. Therefore generally Cr, V, Zn and Y decrease in granitic rocks during differentiation as well as Ni when detected (Tables 1, 2 and 3). However Li generally increases in granitic rocks during differentiation, which is attributed to the fact that the increase in Li content of muscovite is higher than that of coexisting biotite during differentiation in those series and the amount of muscovite increases.

In the tin-bearing granites from Ervedosa and Alijó-Sanfins, the majority of the granite Sn is held in muscovite and it commonly increases with differentiation, mainly because the amount of muscovite increases, while that of biotite decreases.

Sr of granitic rocks will be mainly retained in plagioclase. However K-feldspar is richer in Sr than coexisting albite (Neiva 1977). Micas play a very small role in the Sr content of host granitic rocks.

Rb content of granitic rocks is mainly retained in micas and K-feldspar. Commonly biotite has higher Rb content than coexisting muscovite (Tables 4 to 9), but also similar to higher Rb content than coexisting K-feldspar as found in granites from Alijó-Sanfins (Neiva 1973). The increase in the amount of muscovite through the differentiation series favours the increase in Rb content of granitic rocks during differentiation (Tables 1, 2 and 3).

As Ba replaces mainly K, K-feldspar has a higher Ba content than coexisting micas (Neiva 1973). However, the decrease in the amount of biotite through the differentiation series contributes to the decrease in Ba content of the host granitic rocks.

Conclusions

- The behaviours of Cr, V, Sc, Ba, Nb, Zn, Sn, Li, Rb and Cs in primary biotite (mainly Fe^{2+}-biotite and siderophyllite) and muscovite from nine peraluminous granitic differentiation series are controlled by fractionation of granitic melts.
- Disequilibrium commonly occurs for trace elements between coexisting micas. Regression lines of correlations for trace elements between coexisting micas generally do not pass through the origin due to solid-liquid reequilibrium during late-magmatic evolution.
- Correlations for Cr, V, Nb, Li, Rb and Cs between coexisting biotite and muscovite show different slopes in several series due to different degrees of fractional crystallization and solid-liquid reequilibration during late-magmatic evolution.
- In general, Cr, V, Nb, Zn, Li and Ni prefer biotite rather than muscovite, because they are accommodated in octahedral sites replacing major elements, which have higher contents in biotite than muscovite.
- In unaltered tin-bearing granites, most of the granite Sn is held by muscovite and it increases during differentiation mainly due to increase in the amount of muscovite.
- Biotite has higher Rb and Cs contents than coexisting muscovite, because these trace elements replace K and are accommodated in the interlayer site, which is smaller in muscovite than biotite.
- The partition ratio for Cs in the Serra da Estrela series is 2.8 and similar to that found experimentally.
- Y probably replaces Ca and prefers biotite, while Sr and Ba generally have higher contents in muscovite than coexisting biotite. The behaviour of Sr is supported by experimental data and other studies, while that of Ba is distinct from that of experimental data, but it is in accordance with findings of other authors.
- In granitic rocks, Cr, V, Zn, Sn, Y, Ni and Li contents are mainly retained in micas, it is conect in biotite, while Sr is concentrated in plagioclase and Rb and Ba occur mainly in K-feldspar, but also in micas.

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